(19) Europäisches Patentamt
European Patent Office
Office européen des brevets

(11) Publication number : 0 633 049 A1

(12) **EUROPEAN PATENT APPLICATION**

(21) Application number : 94890057.6

(22) Date of filing : 11.03.94

(51) Int. Cl.[6] : B01D 43/00, B01J 19/10

(30) Priority : 11.05.93 AT 926/93

(43) Date of publication of application : 11.01.95 Bulletin 95/02

(84) Designated Contracting States : BE CH DE DK ES FR GB GR IE IT LI NL SE

(71) Applicant : Trampler, Felix, Dipl. Ing.
Hauptstrasse 55/A4
A-2371 Hinterbrühl (AT)

(71) Applicant : BENES, Ewald
Klosterstrasse 23
A-2362 Biedermannsdorf (AT)

(72) Inventor : Trampler, Felix
Hauptstrasse 55/A4
A-2371 Hinterbrühl (AT)
Inventor : Benes, Ewald
Klosterstrasse 23
A-2362 Biedermannsdorf (AT)
Inventor : Burger, Wolfgang
Zehenthofgasse 19/5
A-1190 Wien (AT)
Inventor : Gröschl, Martin
Praterstrasse 35/18
A-1020 Wien (AT)

(74) Representative : Babeluk, Michael, Dipl.-Ing.
Patentanwalt
Albertgasse 10/8
A-1080 Wien (AT)

(54) **Method for treating a liquid.**

(57) Particulate material dispersed in a fluid is separated by means of an ultrasonic resonance field. In a typical arrangement, the ultrasonic resonance field is generated within a multilayered composite resonator system including the dispersion (S) in a vessel, a transducer (T) and a mirror (M). All layers are parallel to each other. The resonance step-up of the acoustic displacement amplitude in the dispersion is made much higher than the resonance step-up of the acoustic displacement amplitude in all other layers. This is reached by matching a composite resonance frequency of the multilayered resonator and the electrical driving frequency, while simultaneously mismatching the Eigenfrequencies of the transducer and the electrical driving frequency. As a consequence, the electrical power needed for the separation process can be dramatically reduced. Generally, the apparatus is suitable for all kinds of dispersions (solid, liquid or gaseous disperse phases) and is especially powerful for hydrosoles (particles in water) and for separation of biological particles (cells).

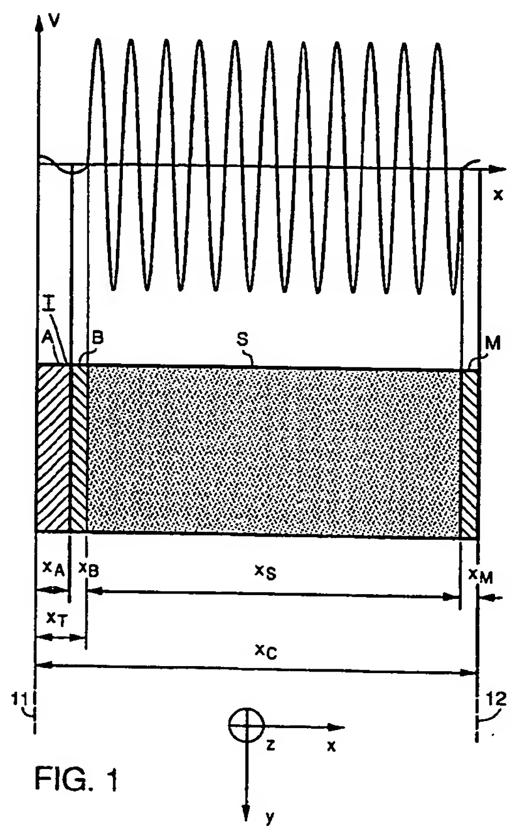


FIG. 1

EP 0 633 049 A1

Field of the Invention

Generally, the present invention is related to a method and to an apparatus to generate ultrasonic resonance fields within a liquid. In more detail the present invention is related to an apparatus for separating particles dispersed in a fluid. It is particularly directed to an apparatus capable of continuously separating dispersed particles with physical properties (especially compressibility, sound velocity, density), different from the fluid, whereby an ultrasonic resonance field is generated within a multilayered composite resonator system including a vessel, a transducer and mirror. Said vessel contains a fluid carrying the particles to be separated. Acoustic radiation force moves the particles towards the nodes or antinodes of the standing wave. Optionally, secondary lateral acoustic forces cause them to aggregate and the aggregates settle by gravity. Numerous fields of modern technology require that particles be removed from fluid. Such separation processes permit either the dispersion medium or particulate matter to be recycled. For example, many industrial processes generate waste water which is contaminated by particulate matter. In Biotechnology, medium has to be separated from biomass. The apparatus is in direct competition to centrifuges and filters, as well as to conventional sedimentation processes using chemical flocculants. The apparatus can also be utilized to sterilize biological matter and for the inactivation of microorganism. In the field of sonochemistry the invention can be applied to accelerate chemical reactions or interactions within the sonicated solution.

Prior Art

Recently, great effort has been directed at the development of acoustic separation or treatment methods to replace or enhance conventional technologies. The establishment of a standing wave in a fluid results in the formation of velocity nodes or antinodes to which the particles are forced to migrate by the radiation force, depending on their compressibility and density. (Most solid and liquid particles move toward the velocity antinodes.) Nodes and antinode planes are at right angles to the direction of propagation of the sound waves, and the nodes are spaced from adjacent nodes by a distance equal to one-half of the wavelength of the acoustic wave in the dispersion. The aggregating effect of ultrasonic sound within those antinodes is prominently known from literature. From E. Skudrzyk, "Die Grundlagen der Akustik", Springer Verlag, Wien, 1954, S. 202-205, S. 807-825; L. Bergmann "Der Ultraschall und seine Anwendungen in Wissenschaft und Technik", Verlag hirzel, Zuerich, 1954: as well as K. Asai and N. Sasaki, "Treatment of slick by means of ultrasonics", Proceedings of the 3rd International Congress on Coal Preparation, Institut National de l'Industrie Charbonniere, Brussels-Liege, 1958, it follows that the frequency to be used in the applied sound is best chosen to be in the range of the so-called characteristic frequency f_o, which can be calculated from

$$f_o = (3\eta)/(2\pi r^2) = 0.48\eta/r^2, \quad (I)$$

whereby η constitutes the kinematic viscosity and r the radius of the particle. Using this frequency range, the effect of radiation force and cumulative acoustically induced Bernoulli forces within the antinode planes can be maximized.

According to the U.S. Pat. No. 4,055,491, ultrasonic standing waves are used to flocculate small particles, such as blood or algae cells, within the velocity antinodes of the acoustic field so that they settle out of the carrying liquid by gravity. But the undefined placement of the ultrasonic source and therefore low efficiency of the standing wave field due to undefined resonance boundary conditions result in high energy losses due to a considerable fraction of traveling waves. It appears that the described process is limited to "batch"-operations since a laminar flow can not be achieved in the preferred embodiments. The apparatus presented in the U.S. Pat. No. 5,164,094 mainly improves laminarity of the flow compared to the embodiments described in U.S. Pat. No. 4,055,491. However, a considerable portion of energy is still lost since frequencies of the sound field applied to the vessels carrying the dispersion is are not controlled by well defined resonance boundary conditions.

An embodiment to separate particles with various acoustic qualities is described in the U.S. Pat. No. 4,523,682. A low resonance mode of a vessel containing a dispersion is excited by a relatively small transducer mounted at one end of the vessel, resulting in node and antinode planes perpendicular to the transducer/vessel interface. Perpendicular modes created by the acoustic point source mean that the system cannot be described as a one-dimensional resonator. The fraction of attenuated traveling waves in the longitudinal direction is high compared to the accumulated acoustic energy in the transversal standing wave field. Acoustic attenuation results in a temperature increase within the dispersion along the flow direction. Temperature changes affect sound velocity and resonance frequency, and cause a non homogeneous temperature distribution along the flow direction which decreases the resonance quality of the field. As a result, the treatment period necessary to achieve desired separation is prolonged.

Because of the long acoustic treatment periods necessary to achieve aggregation and sedimentation of

the particles captured in the antinode planes, efforts were undertaken to move the antinodes of a standing wave relative to the dispersion, in order to obtain the desired separation effect directly by utilizing acoustic forces alone. U.S. Pat. No. 4,673,512 introduces an interference standing wave field generated by opposing transducers which are excited with the same frequency. By controlling the phase shift between the electric excitation signals of the two acoustic sources, it is possible to move particles trapped within the antinodes or nodes of the traveling interference pattern in the dispersion. Using this method, a relatively short treatment period can be achieved. The disadvantage of this method is its non-resonant nature. Much more energy is used to maintain an interference standing wave field compared to a resonant standing wave field of the same amplitude. The result is high electrical power consumption for producing a given acoustic particle velocity amplitude. The same problem has to be considered in U.S. Pat. No. 4,759,775, in which only the method of creating the traveling interference pattern is different.

U.S. Pat. No. 4,877,516 introduces the idea of the controlled movement of local gradients of the acoustic amplitude of the standing field perpendicular to the direction of sound propagation. Thus, particles are moved within the antinodes or nodes of the field by the Bernoulli-force which is directly related to described gradients and is acting parallel to the antinode planes. The disadvantage of the embodiment is the requirement of mechanically moving array to produce acoustic shadows in order to achieve the desired movement of local gradients of the standing wave.

Stepwise movement of the antinodes of a resonant standing wave by exciting succeeding resonance modes of the resonator system is described in the PCT Appl. No. PCT/AT89/00098. Although resonance boundary conditions are fulfilled in some of the described embodiments, there is still considerable acoustically induced dissipation due to the resonator frequencies used, which have always been chosen very close to an Eigen-frequency of the transducer in the past.

Definitions

In order to avoid any misunderstanding in comparing the prior art with the object of the invention, the following definitions are strictly used throughout this description.

Acoustic particles are simply the volume-elements of the acoustic continuum theory and must not be confused with the dispersed particles. The *acoustic particle velocity* is the time derivative of the periodic acoustic particle displacement caused by the regarded sound wave.

One-dimensional treatment of composite resonators means, that an approximate model is applied, where all quantities are regarded as being exclusively dependent from only one direction (Compare, e. g.: H. Nowotny, E. Benes, and M. Schmid, J. Acoust. Soc. Am. 90 (3), September 1991). This direction is coinciding throughout this description with the longitudinal direction.

The term *layer* is very generally used. Even the liquid is regarded as layer, because only one dimension of the liquid-volume is essential for composite resonators built according to the invention. This essential resonance frequency-determining dimension is usually, but not necessarily, the thickness dimension of the layer. The x-axis of the used coordinates is always chosen in the direction of this essential layer dimension. Since the y- and z-dimensions of the layers are of no relevance (one-dimensional treatment), simply the term *layer dimension* is used for the essential, resonance frequency-determining dimension of the regarded resonator part.

Active layers consist of piezoelectric material, *passive layers* consist of non-piezoelectric material.

Transducer is in the most simple case a single layer of a piezoelectric material. For many reasons it is advantageous to bond several piezoelectric plates with one or on both sides upon passive, electrically insulating carrier layers. According to the invention, additional transformation (sub)layers may be used. The transducer-layer itself consists in the most general case of a number of solid layers, whereby the piezoelectric layer is contained and the outermost layers are in contact with the surrounding air or the liquid layer(s), respectively.

Phase shift φ is the spatial phase shift of the acoustic particle velocity V:

$$\varphi = 2\pi \cdot f_e \cdot x/v \quad \text{(II)}$$

where f_e is the electrical driving frequency, x is the resonance determining dimension of the regarded layer, and v is the sound (phase) velocity of the regarded layer. The total phase shift of a multilayer is the sum of the phase shifts of each layer and the additional phase jumps at the interface planes between adjacent layers with different specific acoustic impedances.

Harmonic Eigen-frequencies of a single layer. The harmonic resonance frequencies or harmonic Eigen-frequencies f_i of a layer are defined by

$$f_i = i \cdot (c/\rho)^{1/2}/(2 \cdot x) \quad \text{(III)}$$

where i is the number of the regarded harmonic frequency, x is the resonance determining dimension, c the effective elastic constant, and ρ the mass-density of the layer.

If the layer is consisting of an electroded piezoelectric material, the effective elastic constant c in equation (III) depends upon the electrical load between the electrodes. For the limit cases of short-circuited and open-loop electrodes, the so called series or parallel Eigen-frequencies are determined, respectively. Only an odd subset of the Eigenfrequencies f_i can be electrically excited.

The *half value bandwith* of an Eigen-frequency or resonance frequency is a frequency-range determined by a lower frequency and a higher frequency at which the electrical active power consumption of the regarded layer (or the multilayered structure respectively) is half the value of the active power consumption of the regarded Eigen- or resonance frequency.

Quasi-harmonic Eigen-frequencies of a multilayer (e.g. a multilayered transducer). While the overtone-frequencies of a single homogeneous layer are integer multiples of the fundamental Eigen-frequency, the overtone-frequencies of a composite resonator are in general not that trivially spaced. For that reason, the Eigen-frequencies of a one-dimensional composite resonator are sometimes called "quasiharmonic resonance frequencies". (Compare, e. g.: E. Benes, J. Appl. Phys., Vol. 56, No. 3, 1 August 1984). However, in the case of one layer with dominating dimension (dispersion layer), in a first approximation also the high overtone composite frequencies may be regarded as equidistant. Since a transducer according to the invention usually consists of more than one single layer, it is a multilayered resonator itself. For such a resonator, the Eigen-frequencies can be defined as the frequencies for which the phase shift φT of the acoustic particle velocity amplitude along the dimension x_T of the transducer between the outermost planes is equal to an integer multiple n of the number π. Not all of these mechanically possible resonance frequencies are piezoelectrically excitable. The excitability depends on the displacement curve along the active layer alone. If this curve is a symmetric one, the transducer is not excitable at the corresponding frequency. This definition of the electrically excitable Eigen-frequencies of a multilayered transducer corresponds with the measurable resonance frequencies, if the transducer is surrounded by vacuum (or air) and the frequency of the driving electrical power generator is tuned to the relative maxima of the electrical active power consumed by the resonator. If the voltage amplitude U_e of the driving power generator is kept constant (very low electrical source impedance), the so called series resonance frequency of the composite structure is determined. If the current amplituce I_e of the driving power generator is kept constant (very high electrical source impedance), the so called parallel resonance frequency of the composite structure is determined.

Longitudinal direction means the direction of the layer dimension. The longitudinal direction coincides with the propagation direction of the sound wave excited by the transducer layer. According to the present invention, resonance modes of the composite resonator are excited in longitudinal direction. Therefore, the direction of the standing resonance wave is referred to as longitudinal direction.

Transversal directions are directions perpendicular to the longitudinal direction. These directions fall in the particle velocity node and antinode planes.

Specific acoustic impedance Z is the acoustic impedance per cross sectional area of the regarded material.

Objects and Description of the Invention

According to the preferred embodiments, the multilayered composite resonator system consists of a plane transducer, a vessel containing a liquid to be treated and a plane acoustic mirror. All acoustically coupled layers are arranged in longitudinal direction and their surfaces are parallel to each other. The transducer may consist of a piezoelectrically active layer, such as PZT (Lead-Zirconate-Titanate) ceramics or Lithium-Niobate monocrystals or PVDF layers, and a solid passive layer acting as carrier of the active layer. The invention is suitable to sonicate solutions and dispersions in order to accelerate physical interactions, biological processes, or chemical reactions between their containts and especially to separate dispersed particles of sizes ranging from 10^{-3} to 1 mm. Preferred resonance frequencies according to acoustic and geometric properties of the particles range from 0.1 to 10 MHz.

The main object of the invention is to provide an ultrasonic resonance field within the liquid layer of a multilayered composite acoustic resonator, capable of separating and recycling particles from fluid or of conducting other treatments, while minimizing electrical power consumption and temperature increase caused by acoustically induced dissipation. Many potential applications for ultrasonic separation processes, especially in biotechnology, require a separation method, where temperature increase is negligibly small in order to avoid thermal damage to the particles. Furthermore, the accumulated energy of the established acoustic resonant field and therefore the the acoustic treatment time required for the desired acoustically induced physical or chemical process depend on a homogeneous temperature distribution. This is because the wavelength of an acoustic wave depends on temperature; undesired spatial temperature gradients in transversal directions result in a non-homogeneous distribution of the total phase shift of the acoustic wave. Defined and constant total phase shift distribution in transversal directions of the composite resonance system is a boundary condition

for maintaining high quality resonance fields in longitudinal direction.

As a result of the present invention, the loss characterizing figure $R = W_e/E_S = P_e \cdot \tau_{Cj}/E_S$ is minimized. The loss figure R is hereby defined as ratio between the active electrical energy consumption W_e (per period $\tau_{Cj} = 1/f_{Cj}$) of the composite resonator system and the reactive accumulated acoustic energy E_S of the resonance field in the fluid; f_{Cj} is the excited quasi-harmonic resonance frequency of the composite resonator. P_e describes the active (root mean square) electrical power input,

$$P_e = \tfrac{1}{2} U_e I_e \cos\varphi, \quad \text{(IV)}$$

whereby U_e, I_e are the amplitudes of the driving voltage and current, respectively, φ is the phase between both.

The accumulated acoustic energy E_S is directly related to the acoustic forces acting on the particles, whereby the energy consumption W_e compensates for attenuation of the acoustic field causing thermal dissipation. A small portion of W_e also represents dielectric losses of the transducer, which are of no relevance to the present invention and will not be mentioned further.

We have found that acoustically induced thermal dissipation of transducer and mirror can be minimized by exciting an acoustic quasi-harmonic composite resonance frequency f_{Cj} of the total resonator, but simultaneously mismatching the driving frequency f_e with any of the electrically exiteable quasiharmonic Eigen-frequencies f_{Ti} of the transducer as well as with any of the Eigen-frequencies f_{Mk} of the mirror. As a consequence, the characterizing loss figure R is significantly reduced. This mismatching between driving frequency and Eigenfrequency of the transducer is not at least obvious, since in the mismatched case the composite resonator shows a rather poor electrical behaviour. E. g., in the locus of electrical admittance curves, the composite resonances, which are represented by circles, are much less recognizeable. The resonance circles appear to be much smaller and are offset from the real axis. Because of these properties, in the general case it is much more difficult to design an electrical driving electronics for maintaining a resonance excitation offside a transducer Eigen-frequency. For that reason, in the past the excitation of a composite resonator has been performed always close to the fundamental or third quasiharmonic Eigen-frequency of the transducer.

Composite resonance frequencies f_{Cj} result from the boundary condition at the terminating, total reflecting surfaces of a composite resonator for a standing wave, whereas the maximum of the acoustic particle velocity amplitude has to coincide with this terminating planes. Therefore, the total phase shift ϕ_C across the total length x_C of a multilayered composite resonance system in longitudinal direction, including all layers, has to be an integer multiple of the number π. Mismatching between the electrical driving frequency f_e and the Eigen-frequencies f_{Ti} of the transducer layer can be obtained for a given driving frequency either by proper chosing the transducer thickness x_T and its relative position within the multilayered resonator, or by directly chosing the driving frequency equal to a composite resonance frequency which is sufficiently offset from any excitable Eigen-frequency of the transducer. In the most general case the offset is sufficient if the driving frequency is chosen outside of any of the halfvalue bandwiths of the Eigenfrequencies f_{Ti} of the transducer. In the preferred embodiment of the invention, the offset is sufficient, if it is chosen higher than a certain minimum offset. The minimum offset is equal to 10% divided by the quasiharmonic number i of the regarded Eigen-frequencies f_{Ti} of the transducer:

$$0 < f_e < [0.9\, f_{T1}]\,;\ [1.1\, f_{T1}] < f_e < [(1 - 0.1/2)\, f_{T2}]\,;\ [(1 + 0.1/2)\, f_{T2}] < f_e < [(1 - 0.1/3)\, f_{T3}]\,;\ [(1 + 0.1/3)\, f_{T3}] < f_e < [(1 - 0.1/4)\, f_{T4}]\,;[(1 + 0.1/4)\, f_{T4}] < f_e < \ldots \quad \text{(V)}$$

Introducing this mismatching, the coincidence of the maxima of the acoustic particle velocity amplitude with both outermost transducer planes is avoided. The characterizing loss figure R is optimized, if the thickness x_T and relative position of the transducer layer being chosen with regard to the driving frequency f_e in such a way that a vanishing acoustic particle velocity amplitude V in the interface plane between transducer and the liquid is obtained. In this preferred case, the mismatching between the driving frequency f_e and all the excitable Eigen-frequencies f_{Ti} of the transducer is maximal and the driving frequency f_e is approximately in the middle of one of the allowed intervals defined in equation (V).

Similar rules are valid for the mirror layer. The thickness x_M of the mirror layer has to be properly chosen in order to avoid excitation of its Eigenfrequencies f_{Mi}. The relative position of the mirror layer, however, is fixed as terminating reflecting layer of the multilayered resonator. Generally, the mirror may also consist of several layers.

The transducer layer may form a terminating layer of the composite resonator. Furthermore, the transducer layer may consist of a piezoelectrically active layer, such as PZT (Lead-Zirconate-Titanate, $Pb(Ti,Zr)O_3$) ceramics, or Lithium-Niobate ($LiNbO_3$) monocrystals, or PVDF (Polyvinylidene Fluoride) polymers, and a solid passive layer acting as a carrier of the active layer with defined thickness to achieve low acoustically induced thermal dissipation. Best results can be achieved by using a carrier material with low acoustic attenuation and with a specific acoustic impedance Z_B in the range of or higher than the specific acoustic impedance Z_A of the piezoelectrically active layer. The thickness x_A of the active layer is preferably of a value which causes a phase shift ϕ_A close to or equal to an odd multiple m of the number π. The thickness of the passive layer x_B is preferably

of a value which causes a phase shift ϕ_B being close to or equal to an odd multiple of half of the number π. Using these criteria, the acoustic particle velocity amplitude at the transducer/liquid interface approaches zero and the boundary condition for exciting an Eigen-frequency f_{Tl} of the transducer is optimally avoided. As a result, we have found that energy dissipation of the transducer is minimized. Besides that, for this preferred arrangement, there is no decrease of excitability compared to the case of matching the driving frequency and one of the Eigen-frequency of a transducer.

The transducer layer can also be positioned between two liquid layers. In that case, the thickness x_A of the active layer is preferably of a value which causes a phase shift ϕ_A close to or equal to an odd multiple m of the number π. The thicknesses of the passive layers x_B, x'_B (if any) on each side of the transducer are preferably of values which cause phase shifts ϕ_B φ'_B being close to or equal to an odd multiple n, n' of half of the number π, respectively. Furthermore, the thicknesses of the liquid layers x_S, x'_S and mirror layers x_M, x'_M on each side of the transducer have to be chosen, so that the acoustic particle velocity amplitude at both transducer/liquid interfaces approaches zero and the boundary condition for exciting an Eigen-frequency f_{Tl} of the transducer is maximally avoided. As a result, we have found that energy dissipation of the transducer is minimized. Preferably, the thicknesses x_B, x'_B of the passive layers are equal and the layers are made of the same material.

An odd multiple q of passive sublayers, each of a thickness $x_{B,k}$ (k = 1.....q) causing a phase shift $\phi_{B,k}$ as described above for the thickness x_B of a single passive layer, with alternating high (in the range of the active layer) and low (in the range of the liquid) acoustic impedance, but starting and ending with high ones, is useful to further reduce energy dissipation when this ensemble of sublayers is arranged between the active layer and the liquid layer. Alternatively, low-impedance sublayers can also be formed by a liquid with low acoustic attenuation, optionally circulating in order to control temperature.

Since the frequencies f_{Cj} of the resonance modes of the composite system vary with temperature of the liquid and if applicable the particle concentration it is of significance to compensate the exciting frequency f_e for resonance frequency drifts in order to maintain constant conditions according to the aim of the invention. This can be achieved by controlling the fine tuning of the exciting frequency f_e by an automatic frequency control (AFC) which maintains the active electrical power consumption P_e of the composite resonator at a relative maximum as criterion for resonance.

Another approach to control the exciting frequency f_e towards a preferred resonance frequency f_{Cj} is to provide an additional active layer (e.g. PZT ceramics, Lithium-Niobate monocrystals, or PVDF layers) as mirror or as part of a composite mirror and to utilize the amplitude of the acoustically induced electrical signal at the electrodes of this said active layer as control criterion for maintaining the excitation of the chosen composite resonance frequency f_{Cj}.

Similar to the thicknesses of the layers of a composite transducer, the thicknesses of the piezoelectrically active and passive layers of a composite mirror are advantageously chosen so that the acoustic particle velocity amplitude at the mirror/liquid interface approaches zero and the boundary condition for exciting an Eigen-frequency f_{Mk} of the mirror is optimally avoided. As a result, we have found that energy dissipation of the mirror is minimized for a given acoustic particle velocity amplitude in the fluid.

Analogous to the sublayers of a transducer as described above, sublayers can be introduced in a composite mirror layer. An odd multiple q of passive sublayers of a composite mirror, each of a thickness $x_{B,k}$ (k = 1.....q) causing phase shifts $\phi_{B,k}$ as described above for a transducer, arranged between active layer of the mirror (if any) and fluid layer, with alternating high (in the range of the active layer) and low (in the range of the liquid) acoustic impedance, but starting and ending with high ones, can be used according to the invention to further lower energy dissipation.

Brief Description of the Drawings

FIG.1 shows the interpretation of the composite resonator as multilayered one-dimensional structure and the course of the resulting acoustic particle velocity amplitude, if the layer dimensions are chosen according to the invention,
FIG.2 shows a version of FIG. 1 with the transducer in between,
FIG.3 shows the schematic of a simple resonator example,
FIG.4 shows a composite resonator with a wave guide layer,
FIG.5 shows the cross section of a composite resonator using a totally reflecting mirror as resonator termination,
FIG.6 is a preferred symmetric version of the resonator according to FIG.5,
FIG.7 shows an example of a transducer layer according to the invention,
FIG.8 shows a preferred frequency interval in which the driving frequency should be chosen.

Detailed Description of the Embodiments and Two Typical Dimensionings

In order to emphasize the essential parts of the embodiments, the layers are labeled with alpha-characters, while all other parts are labeled with numbers.

The lower section of **FIG.1** shows a schematic of the essential parts and dimensions of a typical piezoelectric composite one-dimensional resonator. The transducer layer **T** on the left side preferably consists of an active piezoelectric layer **A** and a passive, electrically isolating, carrier layer **B**. The corresponding layer dimensions are x_T, x_A and x_B, respectively. The transducer is acoustically coupled with the liquid **S**; the liquid layer dimension is x_S. Finally, the resonator is completed by the mirror layer **M** with thickness x_M. Since the composite resonator is surrounded by air with a specific acoustic impedance being some order of magnitudes lower than the acoustic impedance of any solid body, the ultimate terminating reflecting planes are the outside planes **11**, **12** of the transducer layer and the mirror layer, respectively. Thus, the total length x_C of the composite structure is defined between these terminating planes. In the upper section of **FIG.1** the spatial course of the acoustic particle velocity amplitude V along the longitudinal direction x is plotted. If the dimensions, the specific acoustic impedance of the layers as well as the electrical driving frequency f_e are chosen according to the invention, the maximum amplitudes in the liquid are, as indicated, much higher than the maximum amplitudes in the other layers. **FIG.1** shows this amplitude relationship only schematically. The quantitative ratio of the maximum amplitude of the standing resonance wave in the liquid to the maximum amplitude in the transducer is usually higher than indicated in **FIG.1**.

The lower section of **FIG.2** shows a schematic of the essential parts and dimensions of a typical one-dimensional piezoelectric composite resonator with the transducer **T** not only coupled to a first liquid layer **S** with the dimension x_S on the right side, but also coupled to a second liquid layer **S'** with the dimension x'_S on the left side. The second liquid layer **S'** may also serve only as coolant or be simply a waveguide layer (e. g. water). The transducer layer **T** preferably consists of an active piezoelectric layer **A** and two passive, electrically isolating, carrier layers **B** and **B'** on both sides of the active layer **A**. The corresponding layer dimensions are x_T, x_A, x_B, and x'_B, respectively. The transducer is acoustically coupled with the liquid layers **S**, **S'**, respectively. Finally, the resonator is completed on each side by a first mirror layer **M** with thickness x_M on the right side, and by a second mirror layer **M'** with the thickness x'_M on the left side. Since the composite resonator is surrounded by air with a specific acoustic impedance being some orders of magnitude lower than the acoustic impedances of any solid body, the ultimately terminating reflecting planes are the outside planes **11**, **12** of the mirror layers **M'**, **M**, respectively. Thus, the total length x_C of the composite structure is defined between these terminating planes **11**, **12**. In the upper section of **FIG.2** the spatial course of the acoustic particle velocity amplitude V along the longitudinal direction x is plotted. If the dimensions, the specific acoustic impedance of the layers as well as the exciting frequency f_e are chosen according to the invention, the maximum particle velocity amplitudes in the liquid layers are, as indicated, much higher than the maximum amplitudes in the other layers.

FIG.3 shows the schematic of a simple resonator. In this example, the piezoelectric layer is represented in y-direction by three piezoceramic plates or discs **A1**, **A2**, **A3** of an equal thickness x_A, arranged side by side and provided with electrodes. The plates **A1**, **A2**, **A3** are acoustically working parallel (in phase excited), while they are electrically connected in series. The plates **A1**, **A2**, **A3** are bonded to a carrier plate **B** (e.g. glass or Al_2O_3-ceramic) with thickness x_B and can be treated in good approximation as one continuous layer **A** with thickness x_A. The flow direction **6**, **7** of the liquid **S** is in direction y. If the apparatus is used for the separation of particels dispersed in a fluid, the dispersed particels are driven by the acoustic radiation forces in longitudinal direction towards the antinode planes of the acoustic particle velocity, where the dispersed particles are agglomerated. The agglomerations are dragged by the gravity forces pointing downwards in direction z (enforced sedimentation by acoustically stimulated coagulation). The carrier plate **B** and the mirror plate **M** simultaneously perform as walls of the liquid vessel. The rectangular cross-sectioned entrance **1** and exit **2** pipes are made tight to the carrier plate **B** and the mirror plate **M** via Viton®-rubber stripes **4** an **4'** respectively. The distance ($x_B + x_S + x_M$) is precisely determined by the distance-rods **5** and **5'** and the flanges **3**, **3'**.

For example, the essential dimensioning of two resonators according to the invention is as follows:

For the separation of biological cells with diameters of the order of 10 μm the appropriate driving frequency f_e is typically around 2 MHz which is signifcantly higher than the characteristic frequency given by equation (I) in order to avoid cavitation. As standard piezoceramic plates are chosen:

Piezoelectrically active layer **A**:

Material:	PZT Lead-Zirconate-Titanate, Pb(Ti,Zr)O3 piezoceramic Hoechst Sonox P4
Mass density:	$\rho_A = 7800\ kg/m^3$
Effective sound velocity for shortened electrodes:	$v_A = 3950\ m/s$
Specific acoustic impedance:	$Z_A = 30.8 \cdot 10^6\ kg/m^2s$
Thickness:	$x_A = 1\ mm$

The fundamental series resonance frequency can be determinded from equation (III): f_A= 1.97 MHz. The series resonance frequency is relevant, because the driving electronics G is assumed to be of the usual low source impedance type. Six (2 x 3) square plates with 25 mm x 25 mm face dimensions are bonded upon a passive layer B (compare also Fig. 7), the thickness value x_B of this passive layer is chosen equal to a standard glass thickness:

Piezoelectrically passive layer **B**:

Material:	Tempax glass
Mass density:	$\rho_B = 2200\ kg/m3$
Sound velocity:	$v_B = 5430\ m/s$
Spec. ac. impedance:	$Z_B = 12 \cdot 10^6\ kg/m^2s$
Thickness:	$x_B = 2.8\ mm$

The resonance frequencies of the two-layer transducer surrounded by environmental air can be measured or calculated (E. Benes, J. Appl. Phys., Vol. 56, No. 3, 1 August 1984). The first four quasiharmonic frequencies are:

$$f_{T1} = 573500\ Hz,\ f_{T2} = 1371400\ Hz,\ f_{T3} = 1958120\ Hz,\ f_{T4} = 2546390\ Hz.$$

According to equation (V) the advantageous intervalls for the driving frequency f_e are:

$$0\ Hz < f_e < 516150\ Hz\ ;\ 630850\ Hz < f_e < 1302830\ Hz\ ;\ 1439970\ Hz < f_e < 1892849\ Hz;\ 2023391\ Hz < f_e < 2482730\ Hz\ ;....$$

The liquid layer dimension x_S depends for instance on the flow rate required and is chosen to be 25 mm:

Liquid layer **S**:

Material:	Hydrosol
Mass density:	$\rho_S = 1000\ kg/m^3$
Sound velocity:	$v_S = 1500\ m/s$
Spec. ac. impedance:	$Z_S = 1.5 \cdot 10^6\ kg/m^2s$
Thickness:	$x_S = 25\ mm$

Mirror layer **M**:

Material:	Tempax glass
Mass density:	$\rho_M = 2200\ kg/m^3$
Sound velocity:	$v_M = 5430\ m/s$
Spec. ac. impedance:	$Z_M = 12 \cdot 10^6\ kg/m^2s$
Thickness:	$x_M = 1.3\ mm$

These Parameters result in resonance frequencies f_{Cj} of the composite resonator with a distance Δf_{Cj} of approximately 26 kHz. Therefore, there are many composite resonance frequencies falling into the advantageous intervalls. E. g., the first resonance frequencies f_{Cj} in the fourth intervall is 2035555 Hz, the second 2061024 Hz, the third 2087140 Hz; thus, the exciting frequency f_e can be chosen for instance to be equal to 2087140 Hz. Since the fundamental Eigen-frequency of the mirror is 2088460 Hz, the selected driving frequency is not sufficiently mismatched to the mirror resonances, as a consequence, the mirror thickness has to be changed, e. g. to 1.8 mm.

FIG. 8 shows the measured resonance spectrum of electrical active power input versus frequency of a

typical resonator. Dimensionings have been chosen mainly according to the example previously described except that the thickness x_B of the passive layer **B** is now 2.7 mm instead of 2.8 mm. As a result the third and fourth Eigen-frequencies f_{T3} and f_{T4} of the Transducer have slightly been increased to 2.035 MHz and 2.620 MHz respectively. Each peak of the plot represents a high overtone resonance frequency of the resonator system; the Eigen-frequency spectrum of the transducer alone can exactly be determined by remeasuring the electrical active power consumption without the resonator volume **S** filled with liquid. The Eigen-frequencies f_{T3} and f_{T4} can also be localized in good aproximation in **FIG.4** by interpretation of the local minima of active power input between each resonance peaks of the complete resonator system as an indicator for the amount of active power consumed by the tranducer if the influence of Eigen-frequencies of the mirror can be neclected. df_{T3} and df_{T4} represent frequency intervals in the neighborhood of the Eigenfrequencies which are usually broader than the half value-bandwiths of the regarded Eigenfrequencies of the transducer. Excitation frequencies are chosen within these half value-bandwiths to drive resonators of the prior art due to the high excitability of resonance frequencies in that area. In contrast to these disadvantageous intervals df_{T3} and df_{T4}, the frequency interval df_e represents the preferred frequency range in which a resonance frequency should be chosen according to the invention and to equation (V). f_{opt} is located in the middle of the preferred interval df_e and indicates the frequency area in which a vanishing acoustic particle velocity amplitude at the interface between transducer and liquid is optimally achieved according to the invention.

A design which is optimal with respect to the object of the invention uses, except the passive layer **B**, the same layers. The passive layer **B** is made of a thickness x_B that produces a node of the particle velocity amplitude V at the interface transducer/liquid. If the driving frequency f_e is selected to be about the Eigen-frequency f_A of the active layer **A**, which guarantees optimal excitation of the composite resonator, the phase shift ϕ_A in the active layer **A** is equal to π. Since there is an antinode boundary condition at the interface plane **11** between active layer **A** and surrounding air, and since the phase shift ϕ_A in the active layer **A** is equal to π, the phase shift ϕ_B in the passive layer **B** must be chosen to be equal to $\pi/2$ or equal to an odd multiple of $\pi/2$ to obtain a node of the particle velocity V at the interface plane between transducer **T** and liquid **S**. Introducing this phase shift $\pi/2$ in equation (II) yields

$$\phi_B = 2\pi \cdot f_e \cdot x_B/v_B = \pi/2$$

and allows the calculation of x_B. If for a more rugged application the result of X_B = 1.2 mm is mechanically to weak, three times the value can also be used.

Piezoelectrically passive layer B:

Material:	Alumina (Al_2O_3) ceramic
Mass density:	$\rho_B = 3780\ kg/m^3$
Sound velocity:	$v_B = 9650\ m/s$
Specific acoustic impedance:	$Z_B = 36.5 \cdot 10^6\ kg/m^2s$
Thickness:	$x_B = 1.2\ mm$

The first two Eigen-frequencies of this transducer are: f_{T1} = 1335150 HZ, f_{T2} = 2866080 Hz.

According to equation (V) the intended driving frequency f_e = 1,97 MHz is now approximately in the middle of the advantageous intervall: 1468665 Hz < f_e < 2722776 Hz.

In this example the optimum thickness for the mirror is determined from (II)

$$\phi_M = 2\pi \cdot x \cdot f_e/v_M = \pi/2;$$

x = 0.692 mm. Since this value is rather to small for a reasonable mechanical ruggedness, three times this value is chosen x_M = 2.07 mm.

Similar to the composite transducer **T**, the mirror **M** may also consist of a active **A** and a passive **B** layer with the same criteria for choosing the thicknesses x_A and x_B of such layers, respectively. The active layer of the mirror provides an electric signal, which can be used to automatically control the exciting frequency f_e towards a preferred composite resonance frequency f_{Cj}.

FIG.4 shows an extension of the composite resonator according to **FIG.3**. In this example, an additional wave guide layer **W**, filled with a low loss liquid (e.g. distilled water), separated by an acoustically transparent wall **F** from the liquid **S**, is inserted. The dimension x_F of the wall **F** is, with respect to the excitation frequency f_e, either small compared to a quarter of the wavelength or equal to the half-wavelength or a multiple of the halfwavelength in that wall material, or the specific acoustic impedance of the wall material is approximately the same as the specific acoustic impedance of the liquid. In the first case, e.g. Saran® or Mylar® foils with a thickness of 10µm are used as wall **F**. In the second case, the wall **F** can be made virtually of any material, but for a material with a specific acoustic impedance close to the specific acoustic impedance of the liquid **S**, the dimension of the layer **F** is less critical. Using the phase-nomenclature, the acoustically transparent layer **F** produces a phase shift ϕ_F of an integer multiple of π. In the third case, a proper material is e. g. TPX (Me-

thylpentene) or ABS (Acrylonitrile Butadiene Styrene). Due to the uncritical effect of its layer thickness and its low attenuation Polysulfon foils have been found to be an advantageous material to form wall layer **F**. The additional wave guide layer **W** serves as a high quality factor resonator part, which removes the very inhomogeneous near field region of the transducer **T** from the treatment zone **S**, thus significantly reducing the potential for acoustic streaming in the liquid **S**. This resonator design version allows also an enhanced cooling and a temperature control of the system by circulating a liquid between the wave guide layer layer **W** and a thermostat. In this case the side walls **8** and **8'** can be equipped with an entrance and exit pipe, respectively. This resonator design version also proofs the applicability of this invention to the so called drifting resonance field (DRF) concept described in a recent patent application (U.S. Appl. No. 474,813 and PCT Appl. No. PCT/AT89/00098). In the case of the DRF separation procedure, the composite resonator is not only driven at one certain harmonic resonance frequency, but is rather switched repeatedly between, e. g. five to twelve, adjacent, closely spaced resonance quasi-harmonic frequencies f_{C_j}. Particles dispersed in the liquid **S** are moved in a stepwise manner as a result of movements of the antinode planes in longitudinal direction x. This allows the splitting of the dispersion exit pipe **2** into two parts, one **7** for the clarified liquid medium, the other **7'** for the liquid medium highly enriched in dispersed particles. To minimize acoustically induced dissipation, the exciting frequencies of the DRF procedure have to be tuned towards resonance frequencies in the neighborhood of preferred resonance frequencies f_{C_j} according to the invention.

FIG.5 shows the cross section of a composite resonator using a totally reflecting retro-reflector **R** as mirror terminating the composite resonator. Said retro-reflector **R** is preferably formed by two plates at right angle to each other. The orientation of the flow of the liquid **S** is preferably chosen opposite to the orientation of the force of gravity and coincides in **FIG.5** with the z-axis. This resonator version is especially advantageous, since there are no side walls engaged which have to be ignored or neglected with respect to their acoustic influence in order to allow a one-dimensional treatment. In contrast to that, a well defined one-dimensional behavior of resonators according to **FIG.3** and **FIG.4** can often only be obtained, if the lateral dimensions of the layers are much higher than the longitudinal ones, which is sometimes not desirable.

The acoustic material parameters of the retro-reflector **R** must be of a value so that the total reflection condition at the interface between liquid **S** and reflector **R** is fulfilled for the tilt-angle α of the reflector being equal 45°. Total reflection at the interface-planes **12'**, **12"** between liquid and retro-reflector disables any acoustically induced dissipation in the reflector **R**. If the medium of the reflector **R** is chosen isotropic having a value of the sound speed for shear waves equal to or higher than 1.41 times the sound speed for longitudinal waves in the liquid **S**, the limit angle for the total reflection condition at the interface between liquid **S** and reflector **R** is exceeded for the tilt-angle α of the reflector being equal to 45°. (In the case the reflector **R** is made from an anisotropic medium, the lower of the two possible shear-wave sound speeds must be equal to or higher than 1.41 times the sound speed for longitudinal waves in the liquid **S**). That is, the excited sound waves are, for the case of neglected losses within the media, totally reflected already at the interface-planes **12'**, **12"** between liquid and reflector. This condition is fulfilled, e.g. for an aqueous liquid and the reflector wall materials Molybdenum, stainless steel and even for the wall material Aluminum. Although the actual sound paths along the distances x_{S12} and x_{S22} are now falling parallel to the y-axis, the total length of any sound path in the liquid is equal:

$$(x_{S11} + x_{S12} + x_{S13}) = (x_{S21} + x_{S22} + x_{S23}) = 2x_S.$$

Thus, a virtual total reflection plane **12** of an equivalent one-dimensional resonator can be defined, whereby the effective layer thickness x_S of the liquid **S** is constant versus lateral directions y and z and all layer dimensions again can be chosen according to the invention.

FIG.6 is a preferred symmetric version of the resonator shown in **FIG.5**. The main advantage of this design is the use of a square cross section tube, whereby the tube walls simultaneously perform as walls for the liquid and as totally reflecting means for the composite resonator. Each of the thicknesses x_B, $x_{B'}$ of the two passive layers **B**, **B'**, as well as the thickness x_A of the active layer **A**, are chosen according to the invention.

FIG.7 shows a more detailed drawing of a composite transducer. The same drawing applies for an active mirror, which includes a piezoelectric layer **A** according to the invention. In the example shown in **FIG.7**, the piezoelectrically active layer **A** is represented by six piezoelectric plates **A1**, **A2**, **A3**, **A4**, **A5**, **A6**,which are of equal thickness x_A, arranged side by side and provided with electrodes. The plates are electrically connected in series to match the electrical impedance of the transducer to the output of the frequency generator **G**. Said generator **G** provides via the clamps **E1**, **E2**, the electrical excitation signal with a frequency f_e. U_e and I_e are the amplitudes of the driving voltage and current, respectively. The plates are bonded upon an electrically insulating and piezoelectrically passive carrier plate **B** of a thickness x_B, such as glass or Al_2O_3-ceramic, and can be treated in good approximation as one continuous layer with a thickness x_A. The electrical connections between the piezoelectric plates are provided by copper films **I1**, **I2**, **I3**, **I4**, and by electrode layers **J1**, **J2**, **J3**, of a thickness x_E, which causes a phase shift ϕ_E of less than 1/16 of the number π. Said electrodes are deposited

onto the surface of the passive carrier layer **B** next to the active layer **A**. The thicknesses x_A and x_B have defined values according to the invention in order to achieve low acoustically induced thermal dissipation. Best results can be achieved if the specific acoustic impedance Z_B of the passive layer is close to or higher than the specific acoustic impedance of the piezoelectrically active layer Z_A. The thickness x_A of the active layer is preferably close to or equal to a value, which causes a phase shift ϕ_A being an odd multiple m of the number π, the thickness x_B of the passive layer **B** is preferably close to or equal to a value, which causes a phase shift ϕ_B being an odd multiple n of half of the number π. An odd multiple q of passive sublayers **B1**, **B2**, **B3**, (or more) each of a thickness $x_{B,k}$ (k = 1.....q) causing a phase shift $\phi_{B,k}$ as described above for a single passive layer **B** is also useful to lower energy dissipation. Said odd multiple of passive layers **Bk** are of alternating high ($Z_{B,k} \geqq Z_A$, k...odd) and low ($Z_{B,k} < Z_A$, k...even) specific acoustic impedance, but starting and ending with high ones in the range of Z_A of the active layer **A**. In a variation of this arrangement of passive sublayers **Bk**, low-impedance sublayers may also be a fluid.

Claims

1. Method for treating a liquid, especially for separating dispersed particles from a liquid by means of applying a resonant ultrasonic sound field in a multilayered composite resonator, said acoustically coupled layers are formed in the propagation direction (x) of the acoustic wave by at least a piezoelectric transducer (T), a vessel containing the liquid (S), and an acoustic mirror (M), whereby said transducer (T) is driven by an electrical power generator (G) with a driving frequency (f_e) within the range of the half-value bandwidths of a characteristic high overtone quasiharmonic composite resonance-frequency (f_{Cj}), characterized in that the driving frequency (f_e) is chosen outside of the half value bandwith of any of the electrically exitable Eigen-frequencies (f_{Ti}) of the transducer.

2. Method of claim 1, characterized in that the driving frequency (f_e) is chosen so that it is outside of any of the intervalls defined between a lower limit value, which is defined as the regarded electrically excitable quasiharmonic Eigen-frequency of the transducer (f_{Ti}) minus the ratio ($f_{Ti}/10i$) of said Eigen-frequency (f_{Ti}) over ten times the quasiharmonic number (i), and a higher limit value, which is defined as said Eigen-frequency (f_{Ti}) plus the ratio ($f_{Ti}/10i$) of said Eigen-frequency (f_{Ti}) over ten times the quasiharmonic number (i).

3. Method of claim 1 or 2, characterized in that driving frequency (f_e) is chosen in such way that the acoustic particle velocity amplitude (V) in the interface plane between the transducer (T) and the liquid (S) is small compared with the maximum amplitudes within the transducer and preferably about zero.

4. Apparatus for treating liquids, especially for separating dispersed particles from a liquid by means of applying a resonant ultrasonic sound field in a multilayered composite resonator, said acoustically coupled layers are formed in the propagation direction (x) of the acoustic wave by at least a piezoelectric transducer (T), a vessel containing the liquid (S), and an acoustic mirror (M), whereby said transducer (T) is adapted to be driven by an electrical power generator (G) with a driving frequency (f_e) within the range of the half-value bandwidths of a characteristic high overtone quasiharmonic composite resonance-frequency (f_{Cj}), characterized in that the thickness (x_T) and relative position of the transducer layer (T) is chosen so that the driving frequency (f_e) is outside of the half value bandwith of any of the electrically exitable Eigen-frequencies (f_{Ti}) of the transducer.

5. Apparatus of claim 4, characterized in that the thickness (x_T) and relative position of the transducer layer (T) is chosen so that the driving frequency (f_e) is outside of any of the intervalls defined between a lower limit value, which is defined as the regarded electrically excitable quasiharmonic Eigen-frequency of the transducer (f_{Ti}) minus the ratio ($f_{Ti}/10i$) of said Eigen-frequency (f_{Ti}) over ten times the quasiharmonic number (i), and a higher limit value, which is defined as said Eigen-frequency (f_{Ti}) plus the ratio ($f_{Ti}/10i$) of said Eigen-frequency (f_{Ti}) over ten times the quasiharmonic number (i).

6. Apparatus of claims 4 or 5, characterized in that the transducer (T) is in contact with the liquid (S) only at one side, the inner side, whereby the outer interface plane of the transducer (T) serves as first terminating reflecting plane (11) and the outer interface plane of the acoustic mirror (M) serves as second terminating reflecting plane (12) and that the transducer (T) preferably is comprised of a piezoelectric solid layer with electrodes, referred to as active layer (A), and a non-piezoelectric layer, referred to as passive

layer (B), whereby

- the active layer is of a thickness (x_A), which causes a spatial phase shift (ϕ_A) of the acoustic particle velocity amplitude (V), said phase shift (ϕ_A) being close to equal to an odd multiple (m) of the number Pi (π),
- the passive layer (B) is of a thickness (x_B), which causes a spatial phase shift (ϕ_B) of the acoustic particle velocity amplitude (V), said phase shift (ϕ_B) being close to or equal to an odd multiple (n) of half of the number Pi (π), and
- the passive layer (B) is made of a material with a specific acoustic impedance Z_B preferrably close to, or higher than, the specific acoustic impedance Z_A of the active layer (A).

7. Apparatus of claims 4 or 5, characterized in that the transducer (T) is in contact with the liquid (S) at both sides and two separate mirrors (M, M') are terminating the resonator, whereby the outer interface plane of the first mirror (M) serves as first terminating reflecting plane (11) and the outer interface plane of the second mirror (M') serves as second terminating reflecting plane (12) and that the transducer preferably is comprised of one piezoelectric solid layer with electrodes, referred to as active layer (A), and two non-piezoelectric layers, referred to as passive layers (B, B'), whereby
   - the active layer (A) is placed in between the two passive layers (B, B'),
   - the active layer (A) is of a thickness (x_A), which causes a spatial phase shift (ϕ_A) of the acoustic particle velocity amplitude (V), said phase shift (ϕ_A) being close to or equal to an odd multiple (m) of the number Pi (π),
   - the first passive layer (B) is of a thickness (x_B), which causes a first phase shift (ϕ_B) of an odd multiple (n) of half of the number Pi (π), and the second passive layer (B') is of a thickness (x'_B), which causes a second phase shift (φ'_B) of an odd multiple (n') of half of the number Pi (π), whereby the thicknesses (x_B, x'_B) of the passive layers (B, B') have preferably the same value, and
   - said passive layers (B, B') are made of a material with a specific acoustic impedance Z_B preferrably close to, or higher than, the specific acoustic impedance Z_A of the active layer.

8. Apparatus of one of claims 4 to 7, characterized in that the mirror includes a piezoelectric solid layer with electrodes, referred to as active layer (A), and the amplitude of the electrical signal (U_e) produced between the electrodes of the active layer (A) is used as criterion for controlling the exciting frequency (f_e) of the electrical power generator (G) towards the value of a quasiharmonic composite resonance frequency (f_{Cj}) in that the amplitude of the electrical signal (U_e) is maintained at a maximum value.

9. Apparatus of claim 8, characterized in that
   - said active layer (A) of the mirror (M) is of a thickness (x_A), which causes a spatial phase shift (ϕ_A) of the acoustic particle velocity amplitude (V), said phase shift (ϕ_A) being close to or equal to an odd multiple (m) of the number Pi (π),
   - the active layer (A) of the mirror (M) is acoustically coupled with a non-piezoelectric layer, referred to as passive layer (B), with a thickness (x_B) causing a phase shift (ϕ_B) of the acoustic particle velocity amplitude, said phase shift (ϕ_B) being close to or equal to an odd multiple (n) of half of the number Pi (π), and
   - the passive layer (B) is made of a material with a specific acoustic impedance (Z_B) close to, or higher than, the specific acoustic impedance (Z_A) of the active layer;

   whereby preferably said passive layer (B) of the mirror (M) forms one wall of the vessel.

10. Apparatus of one of claims 6 to 9, characterized in that said active layer (A) is formed by a mosaic like structure of piezoelectric plates (A1, A2, A3, A4, A5, A6) of circular, rectangular or quadratic shape and of identical thickness (x_A) bonded upon the piezoelectrically passive layer (B).

11. Apparatus of claim 10, characterized in that said passive layer (B) is an electrically insulating dielectric layer and the electrodes of the piezoelectric plates (A1, A2, A3, A4, A5, A6) are at least in part electrically connected in series by connecting electrode layers (J1, J2, J3) which are deposited onto the passive layer (B), said connecting electrode layers (J1, J2, J3) are made of a thickness (x_E) corresponding to a spatial phase shift (ϕ_E) of the acoustic particle velocity amplitude (V) smaller than 1/16 of the number Pi (π).

12. Apparatus of one of claims 6 to 11, characterized in that said piezoelectrically passive layer (B) consists of an odd multiple (q) of passive sublayers, whereby
    - the thickness ($x_{B,k}$ [k = 1.....q]) of each sublayer causes a spatial phase shift (ϕ_B) of the acoustic particle velocity amplitude (V), said phase shift (ϕ_B) being close to or equal to an odd multiple (n_k) of

half of the number Pi (π), and

- said odd multiple of passive layers are of alternating high and low acoustic impedance, but starting and ending with high values preferrably close to or higher than the specific acoustic impedance (Z_A) of the active layer (A).

13. Apparatus of one of claims 4 to 12, characterized in that the fine tuning of the exciting frequency (f_e) of the electrical power generator (G) towards the value of the quasi-harmonic composite resonance frequency f_{Cj} is made by an automatic frequency control (AFC), which utilizes the root mean square power consumption (P_e) of the composite resonator as criterion for controlling, whereby the root mean square power consumption (P_e) is maintained at a relative maximum value.

14. Apparatus of one of claims 6 to 13, characterized in that an additional wave guide layer (W), consisting of a fluid, is placed between the passive layer (B) of the transducer (T) and the liquid (S), and such waveguide layer is separated from the liquid (S) by an acoustically transparent wall (F) and that preferably the side walls of the wave guide layer (W) are equipped with an entrance and exit pipe for circulating the fluid.

15. Apparatus of claim 14, characterized in that the tuning of the excitation frequency (f_e) is switched repeatedly between, five to twelve, adjacent, closely-spaced composite resonance frequencies f_{Cj}.

16. Apparatus of claim 14, characterized in that the acoustically transparent wall (F) with thickness (x_F) producing a spatial phase shift (ϕ_F) of the acoustic particle velocity amplitude (V), which is much smaller than the half of the number Pi (π) for the excitation frequency (f_e) or which is approximately equal to the number Pi (π) for the excitation frequency (f_e).

17. Apparatus of claim 14, characterized in that the acoustically transparent wall (F) is made of a material with an specific acoustic impedance (Z_F) which is very close to specific acoustic impedance (Z_W) of the waveguide fluid (W).

18. Apparatus of one of claims 4 to 17, characterized in that

- a totally reflecting retro-reflector (R) forms one wall of the vessel, such retro-reflector (R) being formed by two plates (12', 12") at right angle to each other and tilted by 45° to the direction (x) of sound propagation,
- the acoustic parameters of the material of the retro-reflector (R) being of a value that the total reflection condition at the interface between liquid (S) and reflector (R) is fulfilled for the tilt-angle between the reflector plates (12', 12") and the longitudinal direction (x) being equal 45°, and
- the flow of the liquid being oriented parallel to these plates (12', 12") and perpendicular (z) to the longitudinal direction (x), whereby
- the virtual total reflection plane (12) of the equivalent one-dimensional resonator defines the effective layer thickness (x_S) of the liquid (S).

19. Apparatus of one of claims 4 to 17, characterized in that two symmetric, totally reflecting retro-reflectors (R, R') form opposite walls of the vessel, the acoustic parameters of the material of the retro-reflectors (R, R') being of a value that the total reflection condition at the interfaces between the respective liquids (S, S') and reflectors (R, R') is fulfilled, and the flow of the liquids (S, S') being oriented perpendicular (z) to the longitudinal direction (x), whereby the virtual total reflection planes (11, 12) of the equivalent one-dimensional resonator define the effective layer thicknesses (x_S, x'_S) of the liquids (S, S').

20. Apparatus of one of claims 4 to 17, characterized in that the thickness (x_M)of the mirror (M) is chosen to cause a phase shift (ϕ_M) of the acoustic particle velocity amplitude, said phase shift (ϕ_M) being close to or equal to an odd multiple (n) of half of the number Pi (π).

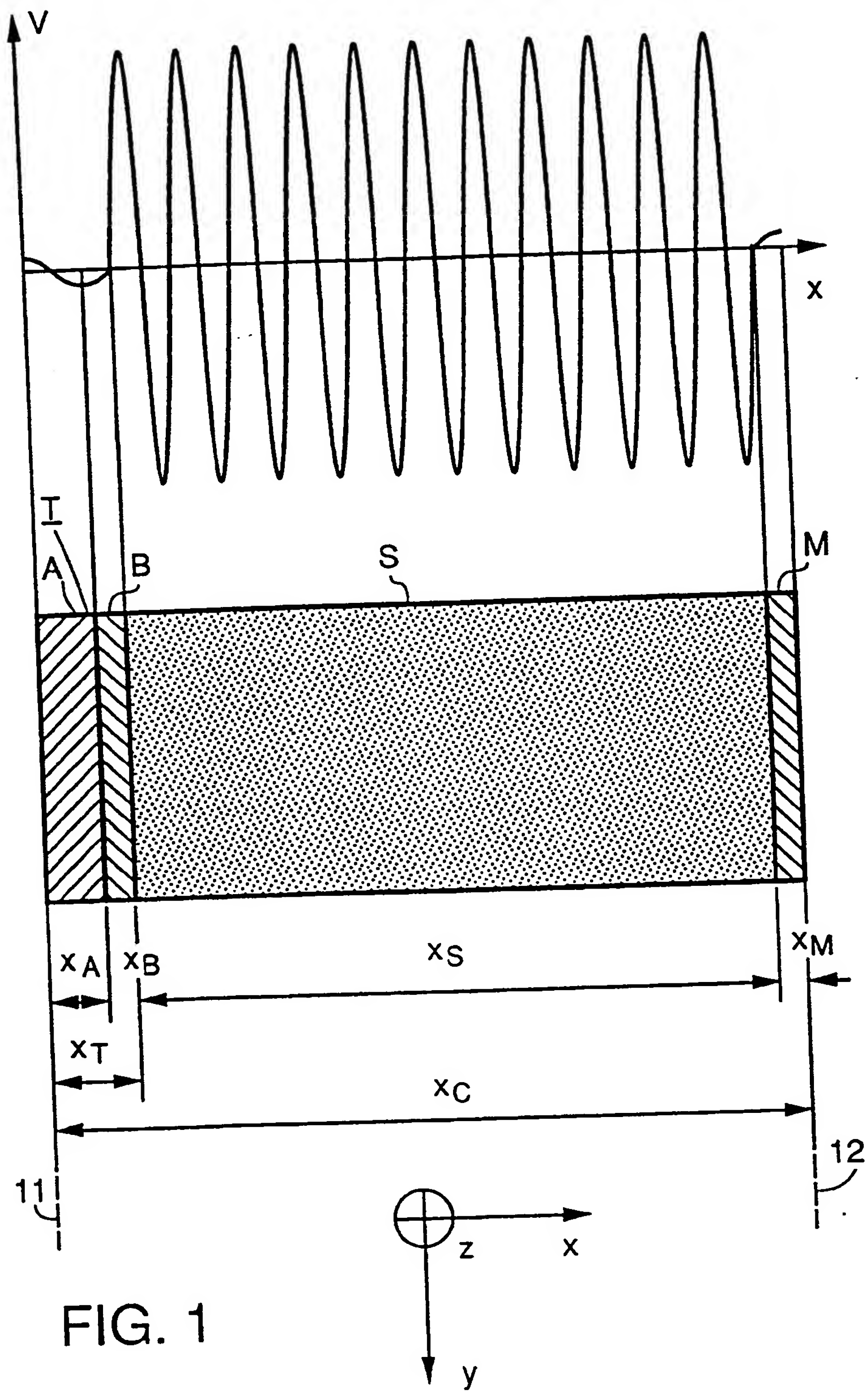


FIG. 1

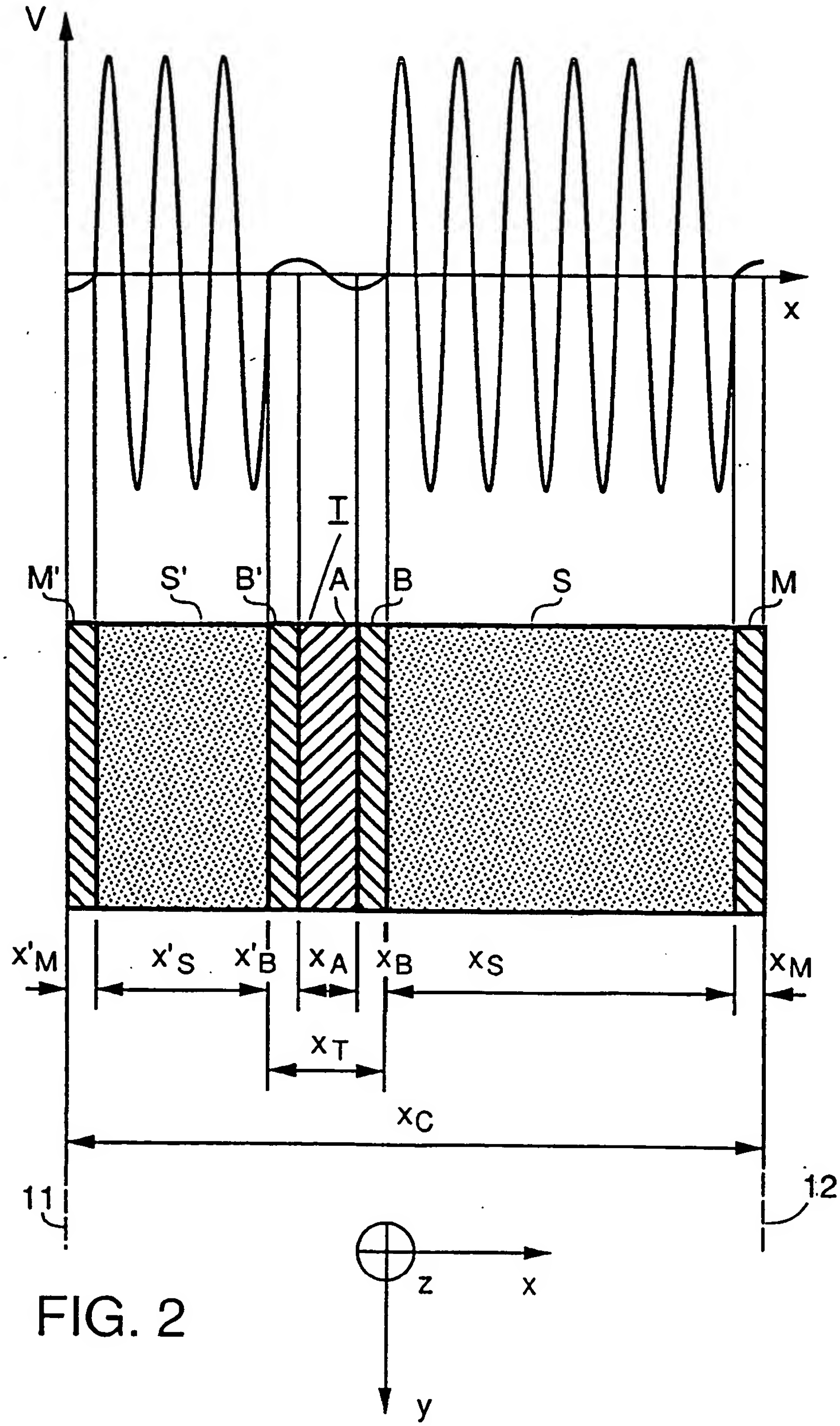


FIG. 2

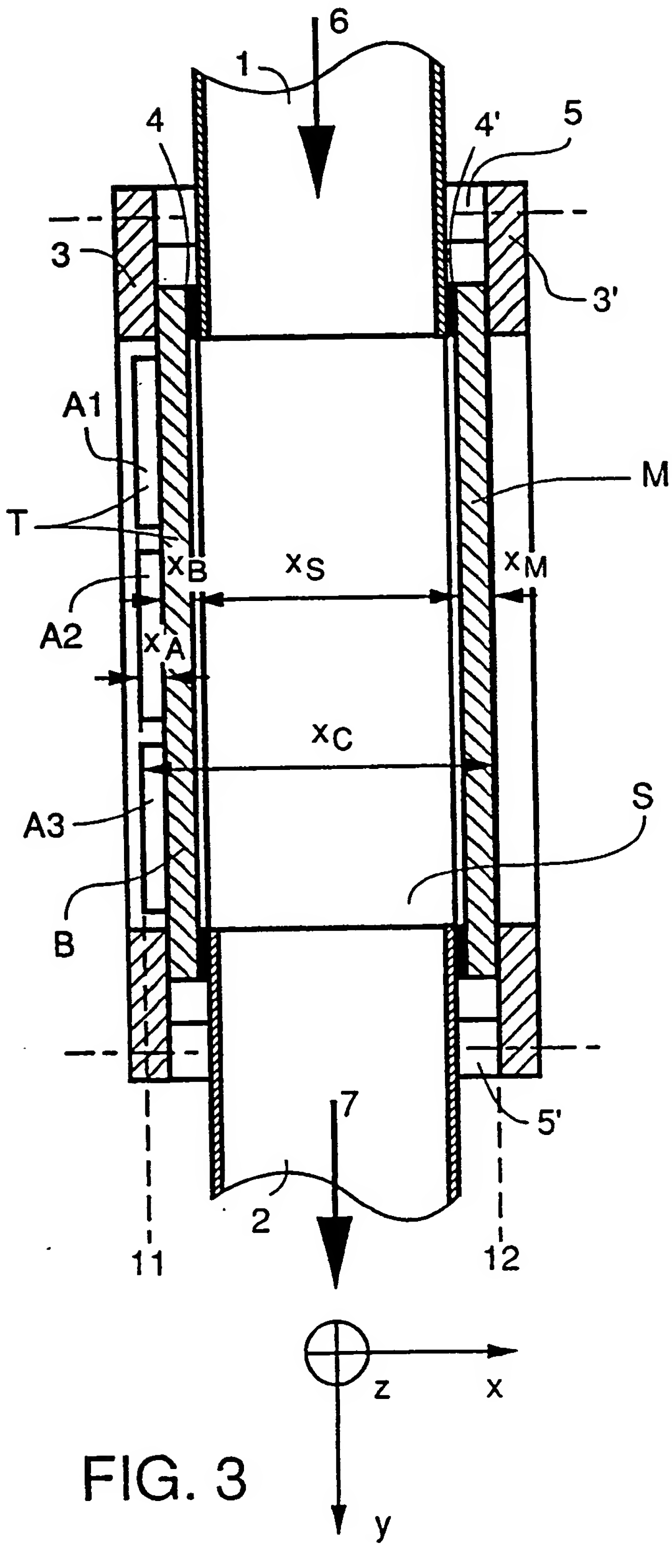


FIG. 3

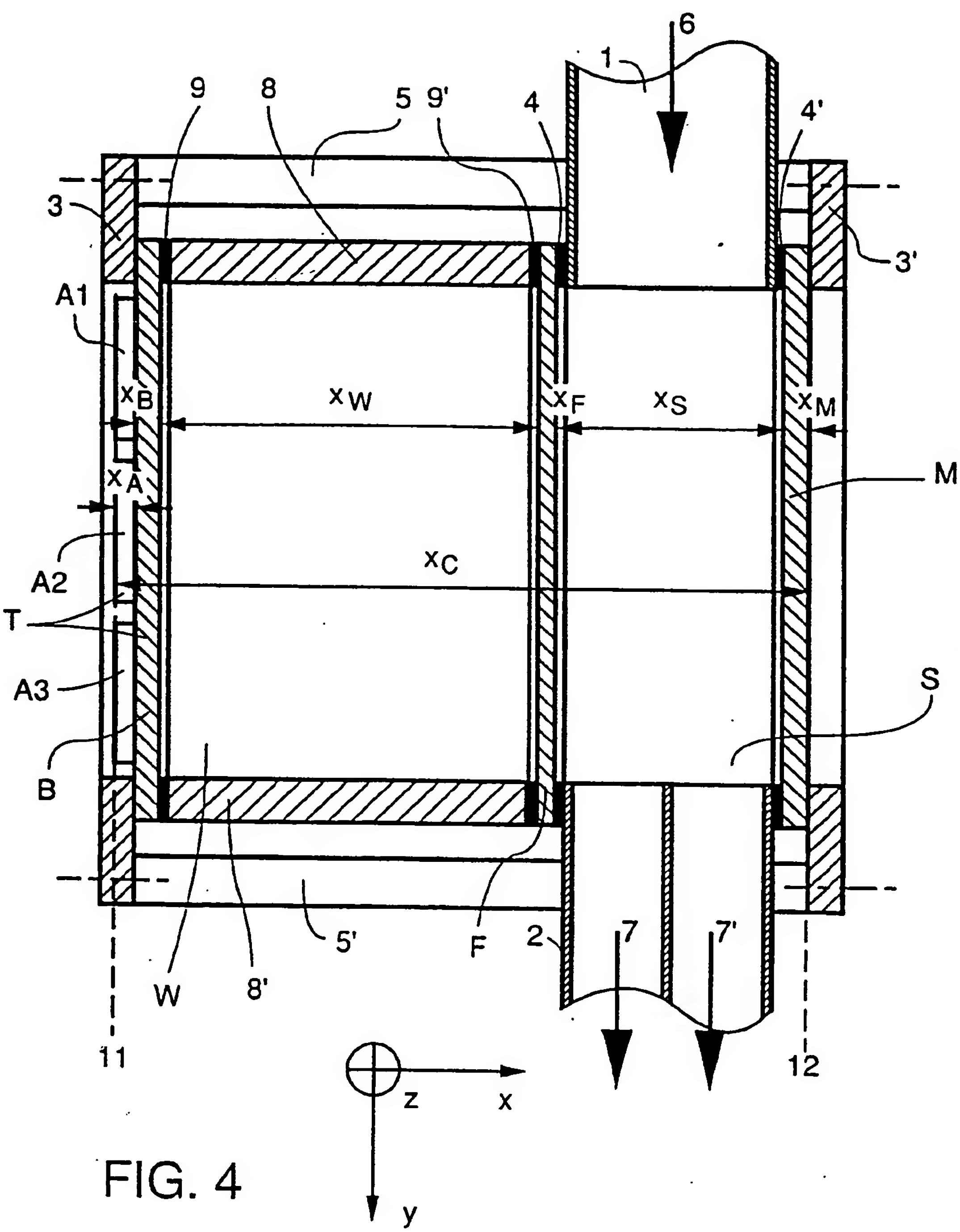


FIG. 4

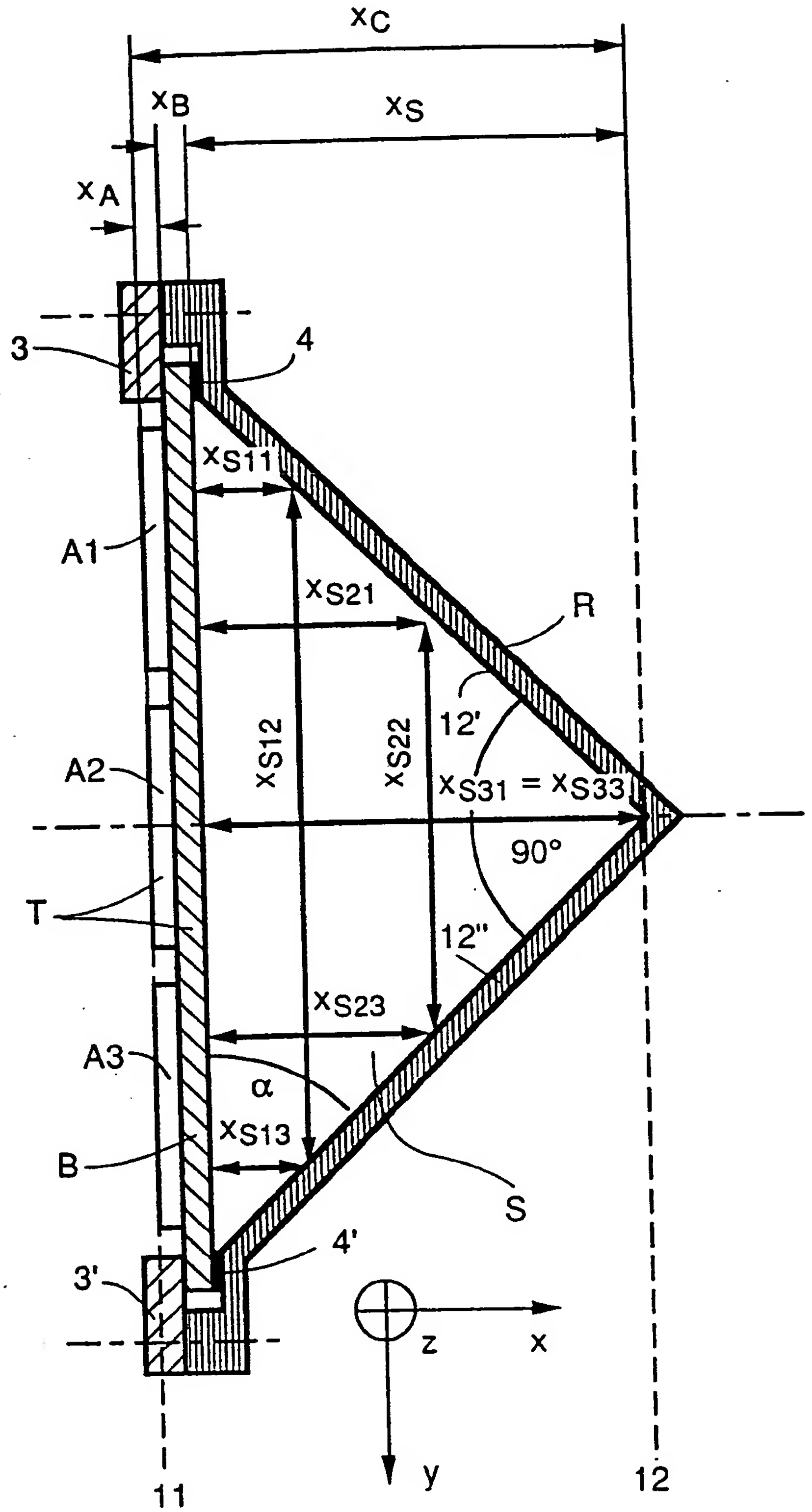

x_C
x_B
x_S
x_A
3
4
x_{S11}
A1
x_{S21}
R
12'
x_{S12}
x_{S22}
A2
$x_{S31} = x_{S33}$
90°
T
12"
x_{S23}
A3
α
x_{S13}
B
S
4'
3'
z
x
y
11
12

FIG. 5

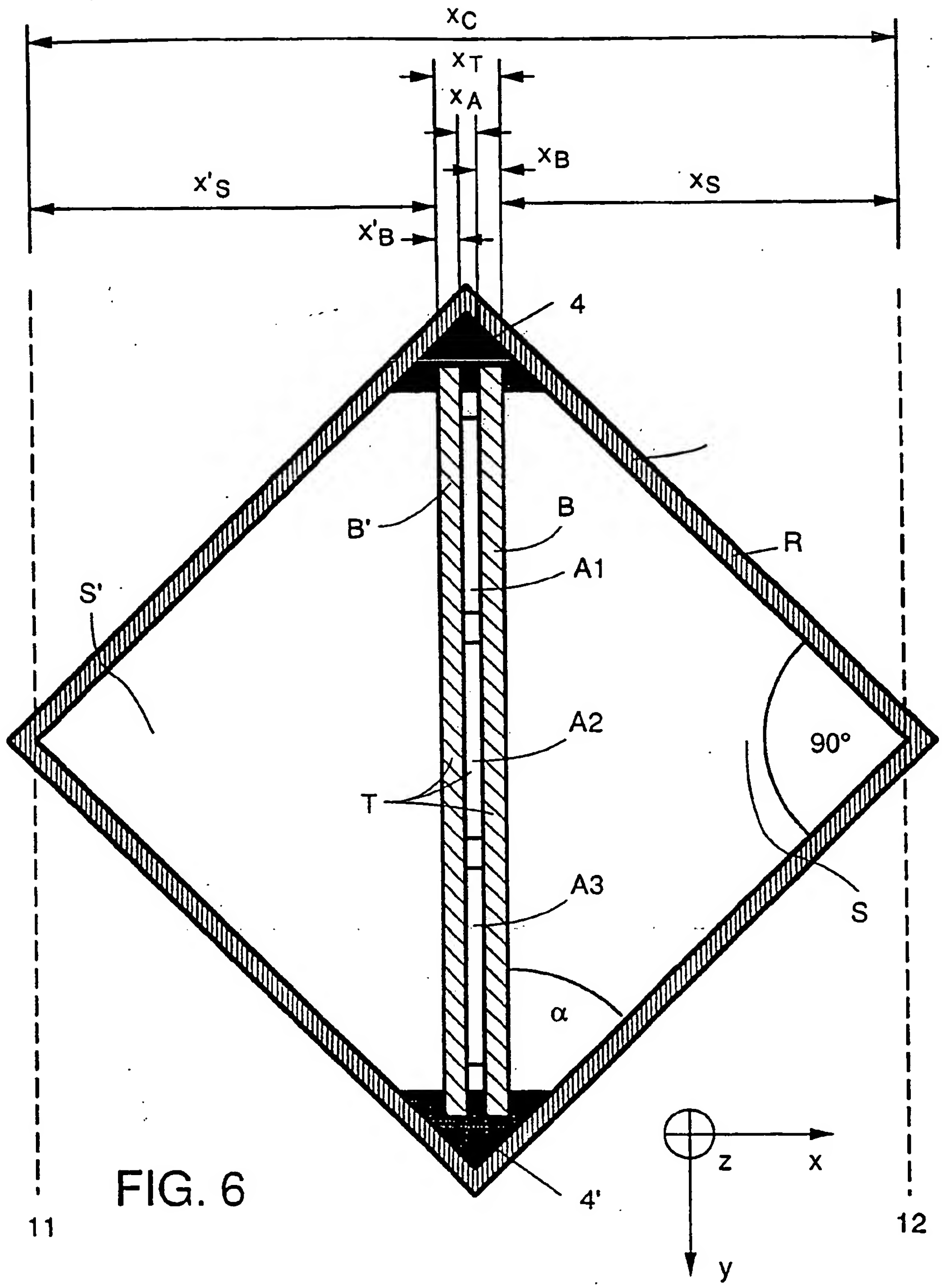
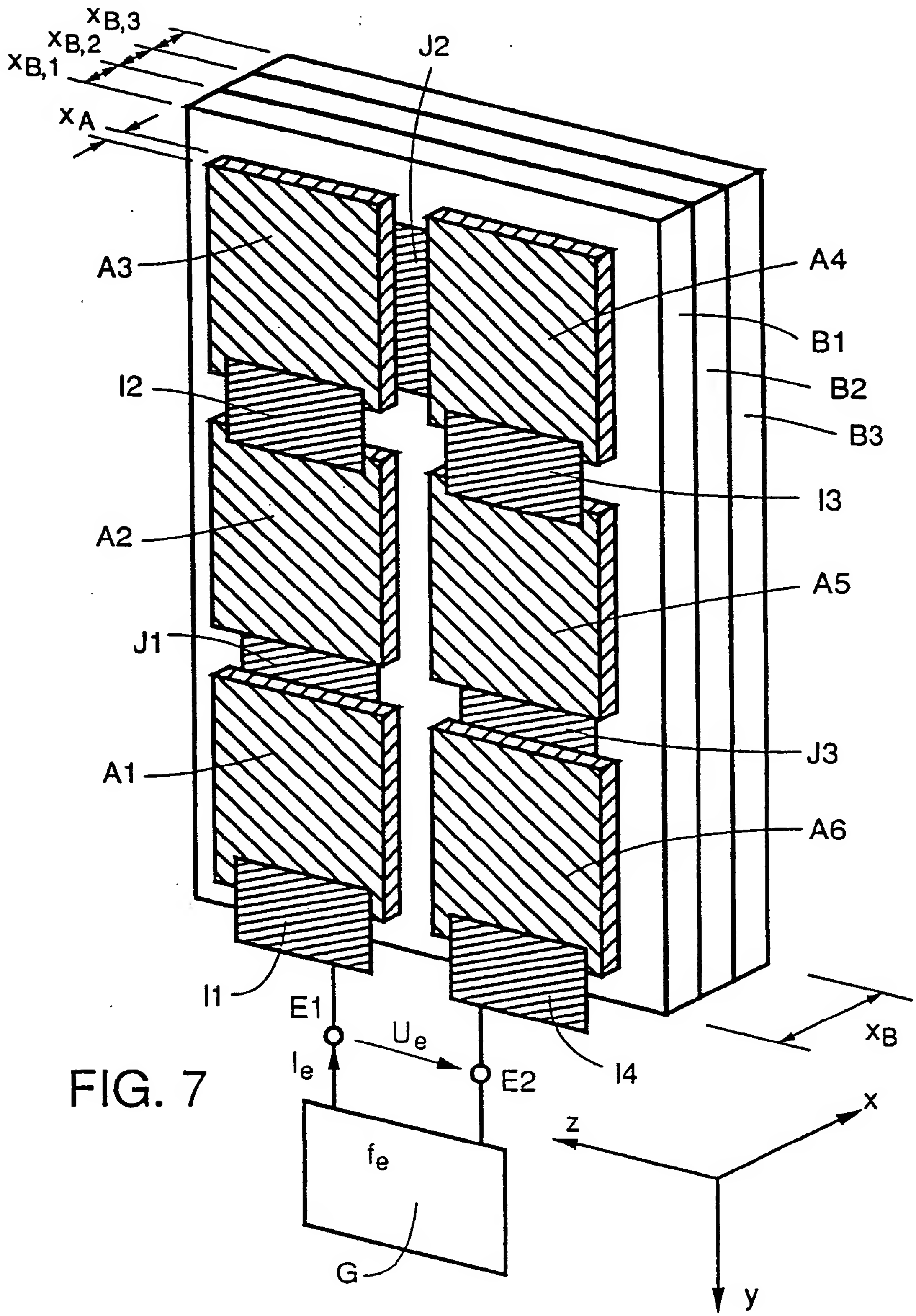


FIG. 6



$x_{B,3}$
$x_{B,2}$
$x_{B,1}$
x_A
J2
A3
A4
B1
B2
B3
I2
I3
A2
A5
J1
J3
A1
A6
I1
E1
U_e
I_e
E2
I4
x_B
x
z
f_e
G
y

FIG. 7

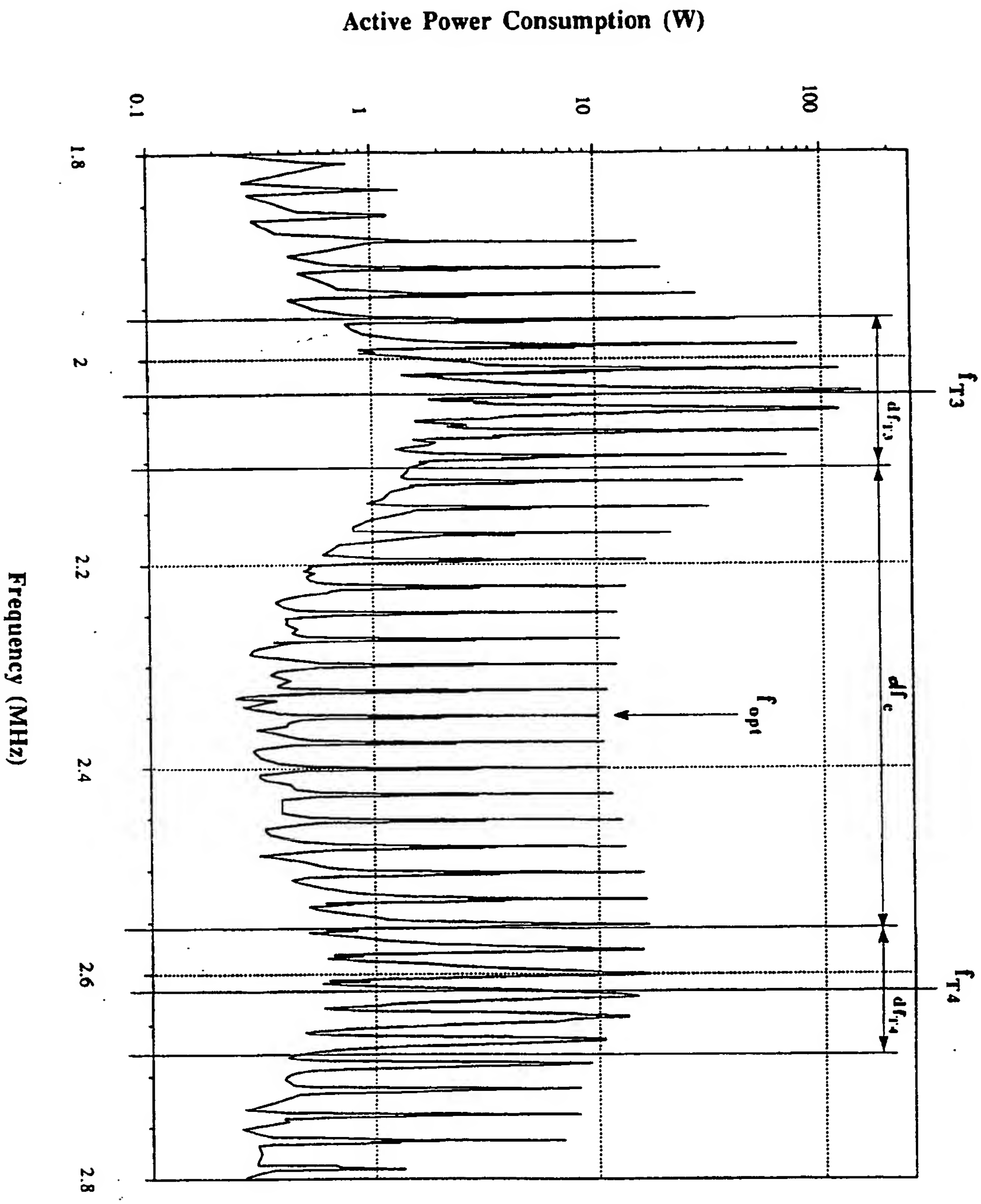

Active Power Consumption (W)
0.1
1
10
100
1.8
2
2.2
2.4
2.6
2.8
Frequency (MHz)
f_{T3}
df_{T3}
df_c
f_{opt}
f_{T4}
df_{T4}

Fig. 8

European Patent Office

EUROPEAN SEARCH REPORT

Application Number
EP 94 89 0057

DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int.Cl.5)
A,D	WO-A-90 05008 (E.BENES ET AL.) * the whole document * ---	1,4,6-9,18	B01D43/00 B01J19/10
A,D	US-A-4 759 775 (S.C.PETERSON ET AL.) * claims 5,15; figure 6 * ---	1,4	
A	EP-A-0 147 032 (UNILEVER PLC) * the whole document * -----	1,4	

TECHNICAL FIELDS SEARCHED (Int.Cl.5)

B01D
B01J

The present search report has been drawn up for all claims

Place of search	Date of completion of the search	Examiner
BERLIN	11 October 1994	Bertram, H

CATEGORY OF CITED DOCUMENTS

X : particularly relevant if taken alone
Y : particularly relevant if combined with another document of the same category
A : technological background
O : non-written disclosure
P : intermediate document

T : theory or principle underlying the invention
E : earlier patent document, but published on, or after the filing date
D : document cited in the application
L : document cited for other reasons

& : member of the same patent family, corresponding document

EPO FORM 1503 03.82 (P04C01)

This Page is Inserted by IFW Indexing and Scanning Operations and is not part of the Official Record

BEST AVAILABLE IMAGES

Defective images within this document are accurate representations of the original documents submitted by the applicant.

Defects in the images include but are not limited to the items checked:

- ☐ BLACK BORDERS
- ☐ IMAGE CUT OFF AT TOP, BOTTOM OR SIDES
- ☐ FADED TEXT OR DRAWING
- ☐ BLURRED OR ILLEGIBLE TEXT OR DRAWING
- ☐ SKEWED/SLANTED IMAGES
- ☐ COLOR OR BLACK AND WHITE PHOTOGRAPHS
- ☐ GRAY SCALE DOCUMENTS
- ☑ LINES OR MARKS ON ORIGINAL DOCUMENT
- ☐ REFERENCE(S) OR EXHIBIT(S) SUBMITTED ARE POOR QUALITY
- ☐ OTHER: ______________________

IMAGES ARE BEST AVAILABLE COPY.
As rescanning these documents will not correct the image problems checked, please do not report these problems to the IFW Image Problem Mailbox.